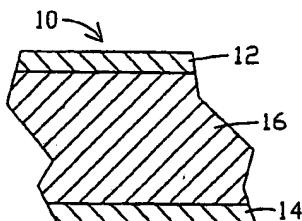




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(54) Title: IN-MOLD LABEL FILM AND METHOD



(57) Abstract

An oriented polymeric in-mold label film (10) comprises a hot-stretched, annealed, linerless self-wound film lamina and has face layer (12) for printing and a base layer (14) which includes a heat-activatable adhesive. The heat-shrinkability of the film (10) is balanced thickness-wise to minimize curl and allow the film (10) to be printed in conventional label-printing presses. An antistat may be included only in the charge for the base layer (14) which includes the heat-activatable adhesive. In the manufacture of labelled blow-molded containers, sheets and labels formed from the film may be handled at high speeds while maintaining accurate registration and dimensional and positional integrity even in the absence of any reinforcing backing, yet the labels perform well on deformable containers such as shampoo bottles.

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IN-MOLD LABEL FILM AND METHOD

2 This application is a continuation-in-part
3 of application Serial No. 07/756,556, filed
4 September 9, 1991.

5 This invention relates to in-mold
6 labelling, using in-mold labels of the kind adapted
7 to label blow-molded plastic containers. Labelling
8 methods and articles of this kind are referred to as
9 "in-mold" because the labels are held in place
10 within the mold, which forms the container during the
11 container-forming step.

12 The invention particularly applies to in-
13 mold labelling using polymeric labels, rather than
14 using paper or paper-like labels. Polymeric labels
15 offer many aesthetic and functional advantages over
16 paper labels in the labelling of containers made by
17 blow-molding plastic resins, such as high density
18 polyethylene (HDPE). When a plastic container such
19 as a HDPE squeeze bottle is used to package a
20 product such as a hair shampoo, a package using a
21 polymeric label is generally more appealing to
22 consumers than a package using a paper label. In
23 many applications the use of polymeric labels is
24 required for reasons of appearance, handling,
25 performance, moisture-resistance, conformability,
26 durability and compatibility with the container to
27 be labelled. Polymeric labels also enable clear or
28 substantially transparent labels with only the label
29 indicia being visible to the consumer.

30 In-mold labelling has significant
31 advantages over methods commonly used in the past to
32 label plastic containers with polymeric labels. The
33 most common of these previous methods involve th

1 use of liner-carried pressure-sensitive adhesive
2 labels, or liner-carried heat-activatable adhesive
3 labels. To produce the liner-carried labels, a
4 laminating step is performed to sandwich a layer of
5 adhesive between a web of label stock and a web of
6 silicone-coated paper which is to function as a
7 carrier or release liner, the label stock is
8 printed, the ink is dried by heating elements or
9 ultraviolet radiation (which also generates heat in
10 the form of infrared), separate labels are cut from
11 the label stock by passing the combination through a
12 rotary-die or flat-bed cutting station, and the
13 matrix of waste or trim label stock (and
14 corresponding excess adhesive) surrounding the
15 formed labels is stripped and discarded or recycled.
16 What remains is a succession of individual labels
17 releasably carried on the release liner.

18 In such earlier methods using carrier-
19 supported polymeric labels, the paper or paper-like
20 carrier or release liner may be relied on to provide
21 dimensional stability to the relatively stretchy and
22 deformable polymeric stock during printing of the
23 labels and ink-drying under heat or ultraviolet, and
24 during die-cutting of the labels and other
25 manipulations which may subject the label stock or
26 labels to mechanical and/or thermal stress on the
27 high-speed printing or labelling lines. This use of
28 the liner to provide dimensional stability avoids
29 distortion of the label stock or labels and
30 resulting interference with continuous high-quality
31 production.

32 In such earlier methods, the labelling of
33 the plastic containers is separate from the
34 manufacture of the containers themselves. At the

1 labelling station, the release liner on which the
2 labels are releasably carried is drawn backwardly
3 around a peel-back edge, thereby deploying the
4 labels one after the other for application to the
5 already-formed plastic containers. The cost of the
6 release liner used in such earlier methods is a
7 significant part of the total material cost
8 associated with labelling, and may even approach the
9 cost of the label stock itself. Therefore, the use
10 of release liner involves a considerable economic
11 cost. Furthermore the liner becomes scrap with
12 little or no reclaim value as soon as it has been
13 employed to dispense the labels. The need to
14 dispose of unreclaimed scrap represents an
15 ecological cost.

16 The matrix of waste or trim label stock
17 involved in such prior methods also involves
18 economic and ecological costs to the extent that the
19 trim (which includes not only label stock but also
20 adhesive) cannot be fully recycled. Even if the
21 trim can be recycled to some extent, the costs of
22 material handling and avoiding contamination
23 incident to recycling also involve real economic and
24 environmental costs.

25 In respect of paper labels, in-mold
26 labelling has been widely practiced for some time.
27 In respect of polymeric labels, in-mold labelling
28 has been proposed as an alternative to the prior
29 methods mentioned above in which release liner or
30 carrier must be used. In-mold labelling using
31 polymeric labels would avoid any use of release
32 liner or carrier, and therefore would avoid the
33 material and ecological costs associated with
34 carrier and matrix disposal or attempted recycling.

1 In in-mold labelling with polymeric labels, self-
2 supported or free-film polymeric label stock (i.e.,
3 linerless polymeric stock) would be combined with
4 heat-activatable adhesive, printed, die-cut and then
5 arranged for deployment, as by being magazine-loaded
6 as a series or stack of linerless labels, or by
7 other means. The polymeric labels would then be
8 sequentially deployed on the molding surface of a
9 blow mold to be bonded onto successive hot
10 workpieces as the workpieces (extruded parisons) are
11 blown and expand against the molding surface and
12 activate the heat-activatable adhesive.

13 Despite the advantages of in-mold
14 labelling over liner-carried labelling, the
15 commercially successful accomplishment of in-mold
16 labelling with polymeric labels has been inhibited
17 by several problems that are not encountered in in-
18 mold labelling using paper labels. One is a lack of
19 an acceptable degree of compatibility with
20 conventional printing presses. The printing of
21 label stock in conventional printing presses used in
22 the label industry subjects the stock to
23 considerable mechanical and thermal stresses
24 incident to the training of the stock through the
25 press and the drying of the ink. Paper stock
26 relatively easily resists these stresses, whether or
27 not the stock is combined with a liner. Also when
28 liner-carried polymeric stock is printed, the
29 dimensional stability of the paper carrier can be
30 relied on to maintain the dimensional integrity of
31 the polymeric stock which tends to stretch and
32 deform under heat. However, there can be no such
33 reliance when free-film polymeric label stock
34 suitable for in-mold labelling is printed. But

1 unless in-mold labelling products and methods are
2 compatible with the use of such conventional
3 printing presses, the threat of obsoleting existing
4 presses presents a strong economic obstacle to wide
5 acceptance of in-mold labelling in the packaging
6 industry.

7 For in-mold labelling with paper labels, a
8 method for die-cutting printed linerless paper label
9 stock into labels and arranging the individual die-
10 cut labels for deployment in the mold involves
11 sheeting the printed label stock, stacking the
12 sheets, forming stacks of individual labels from the
13 stacked sheets with punch dies, and magazine-loading
14 the stacks of individual labels, all done while
15 maintaining proper positioning and registration.
16 The inherent dimensional stability and stiffness of
17 the paper aids in the accomplishment of this
18 process. But labelling with polymeric stock using a
19 similar sequence presents still another problem
20 inhibiting the use of in-mold labelling with
21 polymeric labels-- that of meeting the requirements
22 relating to physical manipulation of the work in
23 process. One requirement is that the linerless
24 printed sheeted polymeric stock be capable of being
25 stacked layer-by-layer in registration so that the
26 die-cutting is accurate. Another requirement is
27 that the individual linerless labels dispense one-
28 by-one in a reliable manner from the magazines and
29 not flutter or deform as they are whipped into
30 position in the mold by high-speed transfer means.

31 The linerless stock and individual labels
32 must meet these requirements, but must also be
33 flexible enough to continue to conform to molded
34 containers to which they are adhered despite flexing

1 or squeezing of those containers.

2 Another problem is that accumulation of
3 static charges must be prevented, since their
4 presence interferes with handling and will prevent
5 accurate stacking and die cutting. The use of
6 antistat agents is known, but their use must not
7 interfere with printing, molding, and label
8 adhesion. The topical application of antistat
9 agents on the face of the label stock after it is
10 printed but prior to sheeting is a possibility but
11 is an expensive and cumbersome step.

12 One example of a recent proposal for in-
13 mold labelling with polymeric labels is found in
14 U.S. Patent 4,837,075 to Dudley. In this patent,
15 polymeric label stock in the form of a multilayer
16 coextrudate (including a layer of heat-activatable
17 adhesive as one of the coextruded layers) is
18 provided which is intended to stand up to handling
19 by high-speed automated equipment during the in-mold
20 labelling procedure. However, Dudley does not
21 address the problem of providing and processing
22 free-film label stock through a conventional label
23 printing press in such a way as to avoid distortion.
24 While he does recognize the importance of avoiding
25 wrinkling or folding during handling by high speed
26 automated equipment, he does not address how to
27 accomplish the same in a manner compatible with the
28 use of conventional printing presses. Nor does he
29 address the problem of eliminating static charges
30 without adversely affecting printing.

31 THE PRESENT INVENTION

32 The present invention overcomes the
33 problems discussed above. The invention

1 contemplates combining a plurality of at least two
2 laminae of film-forming resin to form an oriented
3 polymeric in-mold label film, one of the laminae
4 including a heat-activatable adhesive. The laminae
5 may comprise coextruded layers which are processed
6 together to form the label film, or the laminae may
7 be separately formed laminae and/or lamina which may
8 be combined before, during or after orienting of the
9 film. The film is preferably uniaxially stretched
10 and thereby uniaxially oriented in the machine
11 direction. However, it is contemplated that the
12 film may be stretched in both the machine and cross
13 directions to be thereby biaxially oriented. In
14 such case, the degree of stretch in the machine
15 direction should exceed that in the cross direction
16 so as to give a greater degree of stretch (and
17 stiffness) in the machine direction. The invention
18 involves the concepts, alone or in combination, of
19 (1) hot-stretching or orienting and annealing or
20 heat-setting the coextruded label film or one or
21 more of the label film laminae prior to printing of
22 the stock (and without activating the adhesive even
23 though the temperature of activation of the adhesive
24 is generally lower than the heating temperatures
25 associated with hot-stretching and annealing), (2)
26 thickness-wise balancing of the heat-shrinkability
27 of the extruded layers or laminae so as to minimize
28 curling, and (3) providing an antistat agent in the
29 charge for the adhesive-containing layer or lamina.

30 In a first embodiment, the label film is
31 coextruded and then hot-stretched and annealed. In
32 a second embodiment, non-adhesive laminae or layers
33 of the label film are separately formed and combined

1 with the adhesive-containing layer or lamina before
2 or after hot-stretching and annealing. Unless
3 otherwise indicated by the context of the
4 disclosure, the label film characteristics and
5 preferred properties are applicable to the label
6 films of the first and second embodiments.

7 The invention will be more fully
8 understood from the following more detailed
9 description, taken together with the accompanying
10 drawings, which are highly schematic or diagrammatic
11 and in which FIG. 1 diagrammatically illustrates a
12 coextruded in-mold label film contemplated by the
13 invention; FIG. 2 is a sketch illustrating a
14 coextruding, hot-stretching and annealing line used
15 in the method of the invention; FIG. 3 is a
16 diagrammatic representation of a printing, drying,
17 sheeting and stacking line used in the method of the
18 invention; FIGS. 4-7 diagrammatically illustrate the
19 punch-cutting of in-mold labels into individual
20 stacks of labels in one means of practicing the
21 invention; FIG. 8 diagrammatically illustrates the
22 use of the stacked labels in a molding operation;
23 Fig. 9 diagrammatically illustrates a second
24 embodiment of an in-mold label film having at least
25 one non-coextruded layer; and Fig. 10 is a
26 fragmentary view similar to Fig. 2 showing extrusion
27 coating of the non-coextruded layer of the in-mold
28 label film of the second embodiment.

29 The label film 10 shown in FIG. 1 is a
30 coextrusion consisting of a top or face layer 12,
31 adhesive-containing or base layer 14 which includes
32 a heat-activatable adhesive, and a central or core
33 layer 16. The charges for the several layers are
34 prepared for extrusion through the multifeed

1 coextrusion die 18 as illustrated in FIG. 2. In the
2 particular example described, the total thickness of
3 the film as it leaves the casting station is about
4 20 mils.

5 As is typical with the kinds of polymeric
6 resins and adhesives useful in the practice of the
7 invention, the physical properties of the in-mold
8 label film material are enhanced by hot-stretching
9 and annealing. Hot-stretching is performed at a
10 temperature equal to or above the softening
11 temperature of the film and provides film
12 orientation. Such temperature may exceed the
13 activation or softening temperature of the adhesive.
14 Annealing may similarly involve a processing
15 temperature exceeding the adhesive activation
16 temperature. The in-mold label film material should
17 be annealed at a temperature sufficiently above the
18 expected service temperature to avoid shrinking,
19 relaxing or any distortion of the film which may
20 interfere with the in-mold labelling process. The
21 annealing temperature of the film material is
22 therefore equal to or higher than the temperature at
23 which the heat-activated adhesive is eventually to
24 be activated by contact with the workpieces. In
25 hot-stretching and annealing in the practice of the
26 invention, the extrudate is trained through a series
27 of relatively hot and cool rolls which contact the
28 extrudate to thereby impart heat to and remove heat
29 from the extrudate under time-temperature-direction
30 conditions established by line speed, roll
31 temperature, roll size, and side of contact.
32 According to one important aspect of the invention,
33 the time-temperature-direction conditions are
34 controlled so as to heat at least a majority of the

1 thickness of the extrudate to above its softening
2 temperature prior to stretching without activating
3 the adhesive to an extent that there is sticking of
4 the adhesive to any of the series of heated and
5 cooled rolls that contact the adhesive, and such
6 conditions are also controlled so as to heat at
7 least a majority of the thickness of the extrudate
8 to its annealing temperature following stretching
9 again without activating the adhesive to an extent
10 that there is sticking of the adhesive to any of the
11 series of rolls. This can be successfully
12 accomplished despite the annealing temperature and
13 possibly the softening temperature being equal to or
14 higher than the temperature at which the hot
15 workpieces contact the adhesive to activate it.

16 In accordance with these concepts, in a
17 particular example of the method of the invention,
18 the extrusion die is maintained at 400 degrees F.
19 The extruded film is cast "adhesive side up" (i.e.
20 with layer 14 on the top side and layer 12 on the
21 bottom) onto a casting roll 21 which is maintained
22 at 100 degrees F. and is provided with an air knife
23. The film continues around the casting roll and
24 then passes to the chill roll 22 which is maintained
25 at 70 degrees F. The film continues around the
26 chill roll, trains through the rolls 24, and enters
27 the machine direction orientation unit (MDO unit)
28 25. The film is moved at 15 feet per minute past
29 all these rolls.

30 Within the MDO unit, the film is stretched
31 and stiffened in the machine direction. The film is
32 passed around a first pre-heat roll 26 and then
33 around a second pre-heat roll 28. Both these rolls
34 are maintained at 215 degrees F. and at this point

1 the film continues to move at 15 feet per minute.
2 Although at least a substantial portion of the
3 thickness of the film is heated above its softening
4 temperature so that the hot-stretching operation can
5 be successfully accomplished, the time-temperature
6 relationships are such that the heat-activatable
7 adhesive contained within the adhesive-containing
8 layer is not activated, even though the pre-heat
9 rolls 26 and 28 are at temperatures above the
10 activation temperature of the heat-activatable
11 adhesive. In the particular example described, that
12 activation temperature must be about 200 degrees F.
13 or less, since it must be low enough so at the time
14 the labels are applied in the blow-mold, good
15 adhesion is attained when the layer 14 is contacted
16 by a parison at 200 degrees F. After leaving the
17 second preheat roll 28, the stock tracks on the slow
18 draw roll 31, still moving at 15 feet per minute.
19 The stock then is pulled to the fast draw roll 32
20 which advances the stock at the rate of 75 feet per
21 minute. Therefore, in the particular example
22 described, the stock is stretched fivefold and is
23 drawn down to about one-fifth its original thickness
24 of 20 mils, or to about 4 mils. Stretch ratios of
25 from 2 to 1 to 8 to 1 may be useful in different
26 circumstances, but a ratio between about 4 to 1 and
27 6 to 1 is presently preferred. A midway range
28 extends from about 3 to 1 to about 7 to 1. In the
29 particular example described, the draw rolls 31 and
30 32 are both maintained at 225 degrees F.

31 In the particular example described, the
32 stock now continues on its way at the rate of 75
33 feet per minute. As it leaves the pull-roll pair
34 31,32, the stretched stock is subject to severe

1 shrinkage if it is heated while under little or no
2 mechanical constraint. The plastic stock is said to
3 have a "memory" of its original length to which it
4 tends to return when heated. The stock is cured or
5 annealed to remove this tendency by applying heat to
6 the tensioned stock at the annealing roll 36 which,
7 in the particular example described, is maintained
8 at 240 degrees F. It is to be noted that since the
9 adhesive side is remote from the roll or "up," it
10 does not directly contact the annealing roll 36 and
11 is therefore not directly subjected to the elevated
12 temperature of that roll. The stock then passes
13 directly to the chill roll 38. The roll 38 is
14 maintained at a temperature of 140 degrees F. and is
15 directly contacted by the adhesive-containing layer
16 of the stock. As the stock passes the rolls 36 and
17 38, time, temperature and side of contact all play a
18 part in avoiding activation of the heat-activatable
19 adhesive. After leaving the chill roll 38 at the
20 completion of the hot stretch operation, the stock
21 may be taken up as a self-wound roll 39. The roll
22 39 may be conveniently transported and stored where,
23 as is usually the case, the labels as such are
24 manufactured at a different site than that at which
25 the label stock is manufactured.

26 Such uniaxial hot-stretching of the stock
27 substantially increases stiffness in the machine
28 direction but leaves the stock relatively flexible
29 in the cross direction. As indicated above, it is
30 also contemplated to use unbalanced biaxial
31 stretching of the stock to achieve a satisfactory
32 stiffness differential between the machine and cross
33 directions, with the degrees of stretching and
34 stiffness in the machine direction exceeding those

1 in the cross direction. Whether the stretching is
2 biaxial or uniaxial, that is, whether there is
3 little (relatively) or no stretching in the cross
4 direction, the degree of stretching in the machine
5 direction exceeds that in the cross direction so
6 that the stock is substantially stiffened in the
7 machine direction and remains relatively flexible in
8 the cross direction. Therefore the stock, whether
9 uniaxially or biaxially stretched, may be referred
10 to as having a machine direction stiffness
11 differential. To date, increased stiffness in the
12 machine direction has tended to provide improved
13 labelling manufacture/performance results and no
14 upper limit has been determined. A presently
15 preferred range of stiffness in the machine
16 direction is 40 to 130 Gurley. The cross-direction
17 stiffness tends to be about half or slightly more
18 than half the machine-direction stiffness, say 20 to
19 65. Particularly good results have been obtained
20 with film materials having a machine direction
21 Gurley in the range of 45 to 120 and a cross
22 direction Gurley in the range of 20 to 60. Gurley
23 stiffness is measured in milligrams using the test
24 method designated as TAPPI T543PM-84.

25 Uniaxial hot-stretching and annealing are
26 also important to the development of in-mold label
27 film tensile properties necessary to withstand the
28 mechanical and thermal stresses of conventional
29 printing techniques of the type used in processing
30 paper labels. The stretched and annealed film
31 should have a tensile modulus greater than about
32 65,000 psi and an elongation at break of less than
33 about 950%. Tensile properties including elongation
34 and modulus are measured using the method set forth

1 in ASTM D882.

2 Preferably, the total thickness of the hot
3 coextrudate is about 20 mils, making a total
4 thickness of about 4 mils following hot-stretching.
5 In a presently preferred construction, the face or
6 top layer 12 and the adhesive-containing or base
7 layer 14 each comprise about 10 percent of the total
8 thickness of the hot-stretched film, so that the
9 central layer comprises about 80 percent of the
10 thickness. Alternately, one or both of the face and
11 base layers may be relatively thicker than the
12 central layer.

13 In an example for white (opaque) labels,
14 the layer compositions by weight percentages are:

15 Example 1

16 Top	polypropylene homopolymer	50
17	ethylene-vinyl acetate copolymer	50
18 Central	polypropylene homopolymer	70
19	ethylene-vinyl acetate copolymer	15
20	titanium dioxide concentrate	15
21 Base	heat-activatable adhesive	25
22	antistat	5
23	polypropylene homopolymer	25
24	ethylene-vinyl acetate copolymer	45

25 The different polymer constituents of the various
26 layers comprise physical blends of the indicated
27 polymers with blending being provided in the pellet
28 feed to the extruder and the extrusion process. The
29 titanium dioxide concentrate is itself a blend of
30 50% polypropylene homopolymer and 50% titanium
31 dioxide by weight. The concentrate is available in
32 pellet form for convenience of addition to the
33 extrusion feed.

1 In an example for clear labels, the layer
2 compositions by weight percentages are:

3 Example 2

4	Top	polypropylene homopolymer	50
5		ethylene-vinyl acetate copolymer	50
6	Central	random polypropylene copolymer	60
7		ethylene-vinyl acetate copolymer	40
8	Base	heat-activatable adhesive	25
9		polypropylene homopolymer	25
10		ethylene-vinyl acetate copolymer	45
11		antistat	5

12 The random polypropylene copolymer of the central
13 layer or core contains about 3 to 5% polyethylene by
14 weight. The white label stock is slightly stiffer
15 than the clear label stock. This is believed to be
16 due to the stiffening effect of the titanium
17 dioxide. The stiffness of the clear label stock may
18 be increased by increasing the proportion of the
19 polypropylene to the relatively less stiff ethylene-
20 vinyl acetate ("EVA") or by substituting
21 polypropylene homopolymer for the inherently less
22 stiff polypropylene copolymer. In the following
23 Example 3, the core layer is modified to attain a
24 stiffness similar to that of Example 1.

25 Example 3

26	Top	polypropylene homopolymer	50
27		ethylene-vinyl acetate copolymer	50
28	Central	polypropylene homopolymer	85
29		ethylene-vinyl acetate copolymer	15
30	Base	heat-activatable adhesive	25
31		polypropylene homopolymer	25
32		ethylene-vinyl acetate copolymer	45
33		antistat	5

Example 4

Example 4 is prepared by extrusion of the composition of Example 3 with a thickness of about 22.5 mils to result in a total film thickness of about 4.5 mils after hot-stretching. The ratio of the relative thicknesses of the layers of the film of Example 4 is similar to that of Examples 1-3.

Suitable film-forming polymers for use in the films of the invention are available from a number of commercial sources. The polypropylene homopolymer and copolymer resin materials used herein are sold by Shell Chemical Company under the designations DX 5A97 and 6C20 respectively. The DX 5A97 resin has a melt flow rate of 3.9 g/10 min. (ASTM D1238L), a density of 903 kg/m³ and a flexural or flex modulus of 1,590 MPa (ASTM D790A). The 6C20 resin has a melt flow rate of 1.9 g/10 min. (ASTM D1238L), a density of 895 kg/m³ and a flexural modulus of 806 MPa (ASTM D790A). The ethylene-vinyl acetate copolymer is sold by Quantum Chemical Corp. under the designation UE 631-04. The UE 631-04 resin has a melt flow rate of 2.5 g/10 min. (ASTM 1238E), a density of 940 kg/m³ and a vinyl acetate content of 19% by weight.

The heat-activatable adhesive is a proprietary product sold by H.B. Fuller of Blue Ash, Ohio under product number HM727, and comprises ethylene-vinyl acetate copolymer ("EVA"), polyethylene waxes and a tackifier effective to accomplish adhesion to HDPE. The adhesive is itself far too "watery" or low in viscosity to be successfully extruded, but blends well with the EVA. The EVA stiffens up the extrudate, but is too sticky to process following extrusion, because it tends to

1 stick to processing rolls with which it comes into
2 contact while it is warm so as to damage the
3 adhesive layer or laminate. The addition of
4 polypropylene gives the extrudate excellent heat
5 stability for hot-stretching and other processing.
6 The addition of polypropylene also controls and
7 moderates tackiness so as to make it possible and
8 practical to process the extrudate film according to
9 the concept of controlling time-temperature-
10 direction conditions so as to avoid sticking to the
11 rolls even though activation temperature of the
12 adhesive is below glass transition and annealing
13 temperatures. At the same time, the reduction of
14 tackiness effected by the polypropylene does not
15 interfere with excellent adhesion of the film to a
16 plastic container of HDPE, for example. The
17 proportions of EVA and polypropylene may be varied
18 to accommodate processing variations.

19 The antistat is incorporated in the
20 adhesive-containing or base layer charge and
21 uniformly blended therewith. The amount of antistat
22 used may be varied for particular formulations and
23 processing conditions, the 5% amount used herein
24 being typical. The antistat is efficiently used
25 since it may be added to the adhesive or base layer
26 charge only. Thus, the antistat addition to the
27 base layer charge only provides specificity and
28 efficiency of use without the disadvantages of a
29 topically applied antistat. In certain
30 applications, it may be advantageous to also include
31 the antistat in the central layer charge as well as
32 the base layer charge, or in the central layer
33 charge only.

1 In the particular examples described, the
2 antistat used is sold by Hoechst Celanese under
3 product number E1956 and is of the type that when
4 added in bulk blooms to the surface and dissipates
5 electric charges by hydrophilic action which
6 attracts extremely minute amounts of ambient
7 moisture. By adding the antistat to the adhesive
8 layer only, collection of moisture at the face layer
9 which may interfere with the label printing process
10 is avoided. Most surprising, it has been found that
11 moisture collected at the adhesive layer surface
12 does not interfere with adhesion of the label to the
13 container in the in-mold labelling process. It is
14 believed that the moisture is vaporized or
15 dissipated by the elevated molding service
16 temperatures, but in such small quantities as to not
17 interfere with adhesion.

18 In the following examples, the base layers
19 comprise a blend of polymers found to provide a
20 suitable heat-activatable adhesive for use in
21 connection with polyethylene. In a preferred
22 example for white (opaque) labels, the layer
23 compositions by weight percentages are:

24 Example 5

25 Top	polypropylene homopolymer	50
26	ethylene-vinyl acetate copolymer	50
27 Central	polypropylene homopolymer	70
28	ethylene-vinyl acetate copolymer	15
29	titanium dioxide concentrate	15
30 Base	ethylene-vinyl acetate copolymer	50
31	low density polyethylene	50

1 The composition of the top and central layers of
2 Example 5 correspond with those of Example 1, but
3 the base layer adhesive comprises a 50/50 blend of
4 ethylene-vinyl acetate and low density polyethylene.
5 A suitable low density polyethylene is sold by
6 Rexene Products Company of Dallas, Texas under the
7 designation PE 1017. The PE 1017 resin has a melt
8 flow rate of 2.0 g/10 min. (ASTM 1238E), a density
9 of 920 kg/m³ and a secant modulus of 220 MPa (ASTM
10 638). The remaining polymer and filler components
11 are available as described above.

12 In a preferred example for clear labels,
13 the layer compositions by weight percentage are:

14 Example 6

15	Top	polypropylene homopolymer	50
16		ethylene-vinyl acetate copolymer	50
17	Central	random polypropylene copolymer	80
18		ethylene-vinyl acetate copolymer	20
19	Base	ethylene-vinyl acetate copolymer	50
20		low density polyethylene	50

21 The top layer composition of Example 6 corresponds
22 with that of Example 2, but the central layer
23 proportion of polypropylene copolymer is increased
24 in order to further increase the stiffness. Again,
25 the preferred base layer composition for
26 polyethylene containers is used in Example 6.

27 The antistat is omitted from the
28 compositions of Examples 5 and 6 in favor of the use
29 of an over-varnish applied to the printed label to
30 protect the face side of the label and provide
31 improved slip properties having a reduced
32 coefficient of friction. The reduced coefficient of

1 friction tends to correspondingly reduce the static
2 charge build-up as labels are moved across each
3 other during processing including label stacking and
4 dispensing so as to eliminate or reduce the need for
5 an added antistat agent.

6 It is to be noted that the compositions in
7 the two outer layers are similar. The result is
8 that the construction is well balanced with respect
9 to heat-shrinkability at both sides of the
10 construction. Such balancing of heat-shrinkability
11 is an important concept of the invention.

12 As described above, the presently
13 preferred top and base layer formulations comprise
14 blends of olefin polymers and copolymers of olefin
15 monomers with ethylenically unsaturated carboxylic
16 acid or ethylenically unsaturated carboxylic acid
17 ester comonomers such as the ethylene-vinyl acetate
18 copolymer. The presently preferred central or core
19 layer formulations also comprise blends of olefin
20 polymers and copolymers of olefin monomers with
21 ethylenically unsaturated carboxylic acid or
22 ethylenically unsaturated carboxylic acid ester
23 comonomers such as the ethylene-vinyl acetate
24 copolymer.

25 The composition of the film layers should
26 preferably be compatible with the container
27 composition to enable recycle and regrindability of
28 waste or the like during production. Also,
29 compatible label and container compositions enable
30 post-consumer-use recycling of the container and
31 integral label.

32 As schematically illustrated in FIG. 3,
33 the hot-stretch d stock, which may be supplied in
34 the form of the self-wound roll 39, may be printed

1 or decorated in a printing press 40 in which the
2 stock is subjected to mechanical and thermal stress
3 incident to the printing itself and to the drying of
4 the ink by exposure to heat as such or by exposure
5 to ultraviolet radiation which tends to also
6 generate infrared radiation.

7 Following printing and drying, the stock
8 may be sheeted and stacked in a manner similar to
9 that known for the sheeting of paper-backed label
10 stock. Cutting is indicated by arrow G in the
11 drawings. The severed sheets are stacked to form
12 the stack 44. The stack may contain 100 or 200
13 sheets. For clarity of illustration, in the drawing
14 the thickness of the sheets is greatly exaggerated
15 and the stack 44 is therefore shown as made up of
16 only a relatively small number of sheets. Each
17 sheet in the stack is intended to provide material
18 for several individual labels to be die-cut from the
19 sheeted material. In the particular example
20 described, nine labels are die-cut from each sheet.
21 The sheets in the stack must be very accurately
22 registered with each other so that the labels to be
23 cut from the sheet will be formed in correct
24 registration to the printing that appears on their
25 face according to the pattern printed by the press
26 40.

27 If the linerless unsupported label stock
28 is too limp, accurate stacking is prevented due to
29 the inability to guidingly control positioning of a
30 limp sheet by means of belts, guideways, stops or
31 similar guiding means (not shown) with any degree of
32 accuracy. The stiffening of the linerless stock by
33 uniaxial hot-stretching to desired stiffnesses, as
34 discussed more fully below, allows accurate stacking

1 to be achieved.

2 Accurate stacking and subsequent handling
3 of the sheets or labels formed therefrom is also
4 impeded if static charges are present on the sheets
5 or labels. The antistat present in the base layer
6 acts to remove static charges. In the antistat used
7 in the above examples, this action involves the
8 creation of a very thin layer of moisture at the
9 surface of the base layer. Even though this
10 moisture-containing surface contacts the surface of
11 the hot workpiece as the workpiece is formed, as
12 more fully described below, molding of the workpiece
13 and performance of the adhesive is not adversely
14 affected to any detected degree by flashing of the
15 moisture.

16 Individual labels are formed in a known
17 manner by hollow punches or cutting dies 46 carried
18 on a head 48, seen in bottom plan view in FIG. 4 and
19 in side elevation in FIGS. 5 and 6. The cutting
20 dies punch out the labels from the stack 44,
21 producing in each cutting cycle a number of stacks
22 50 of individual labels. In the particular example
23 described, nine stacks of individual labels are
24 produced in each cutting cycle.

25 Alternatively, following printing and
26 drying, the stock may be fed into a rotary steel die
27 (not shown) at the end of the printing press line
28 and cut into labels. As the cut labels and
29 surrounding matrix of waste material exit from the
30 rotary steel die, the matrix is pulled away at an
31 angle from the labels which are sufficiently stiff
32 to continue their forward travel into a nip of a
33 pair of feed belts (not shown) for collection into
34 stacks 50. Thus, the machine direction stiffness is

1 utilized in a direct label cutting and separating
2 process which eliminates the cutting step at G as
3 well as the other steps described with respect to
4 FIGS. 4, 5 and 6.

5 The stacks 50 of individual labels are
6 stabilized by suitable wrapping or packaging (not
7 shown) in a manner similar to that previously used
8 with paper-backed labels. The stabilized stacks 50
9 are then moved or transported to the site where the
10 blow-molded containers are being manufactured, which
11 often is at a different place than the site of label
12 manufacture.

13 At the site of container manufacture,
14 stacks 50 of individual labels are loaded in a
15 dispensing magazine of a known type, schematically
16 illustrated by magazine 54 in FIG. 8. For example,
17 the labels may be advanced to the front of the
18 magazine by spring means 56, and may be lightly
19 retained for pick-off by springy or mechanically
20 retracting retainer fingers 58. A robotic label
21 feed head 60 carries vacuum cups 62 adapted to be
22 advanced by means (not shown) internal to the head
23 to pick off the front label in the stack 50,
24 retracted for translating movement of the head and
25 the single picked-off label 50a into the opened blow
26 mold 64 by actuation of the translating cylinder 61,
27 and advanced again to apply the picked-off label to
28 the interior surface of the mold and release it.
29 The label may then be held accurately in position
30 within the mold by vacuum applied to the mold wall
31 through vacuum lines 66 while the label feed head 60
32 is retracted. The vacuum line outlets to the
33 interior of the mold may be flush with the interior
34 surface of the mold, as shown, so that the label

1 occupies part of the mold cavity proper. In other
2 words, preferably there is no recess on the interior
3 mold surface to accommodate the label.

4 A hot workpiece or parison (not shown) of
5 polyethylene or similar thermoplastic resin is fed
6 into the mold, the mold is closed, and the parison
7 is expanded in a known manner to complete the
8 formation of the molded container. As the hot
9 parison contacts the adhesive-containing base layer
10 14 of the label, activation of the adhesive is
11 triggered. As indicated above, the annealing
12 temperature of the in-mold label film should exceed
13 the service temperature in the mold in order to
14 avoid label shrinkage or distortion. To assure a
15 uniform joining of the label and container, it is
16 also desirable that the softening temperature of the
17 in-mold label film be close to the service
18 temperature. If, as is preferred, the label is on,
19 not in, the interior surface of the mold, the label
20 becomes embedded in the workpiece to which it is
21 adhered, thus advantageously providing an inset
22 label that is flush with the container surface and
23 that replaces and therefore saves a portion of the
24 charge for the molded workpiece or container without
25 diminishing the structural integrity of the
26 workpiece to any detected degree. As previously
27 indicated, even though the antistat in the base
28 layer causes the presence of moisture at the face of
29 the base layer which must flash off upon contact of
30 the base layer by the surface of the hot parison,
31 the temperature of which may be say about 200
32 degrees F., no detected adverse affect on adhesion
33 or molding occurs.

1 Other mechanisms may be employed involving
2 rotary movement of the robotic parts. Generally,
3 regardless of whether rotary movement is involved,
4 the elements of such label feed mechanisms move at
5 high speed. If the labels are too limp, they tend
6 to flutter, interfering with proper positioning.
7 Also, pick-off becomes unreliable because more than
8 one label at a time may be picked off since limp
9 labels tend to follow each other past the retainer
10 fingers 58. The self-supporting or linerless labels
11 must retain their dimensional and positional
12 integrity without the benefit of a reinforcing
13 backing.

14 Experience to date indicates that a
15 minimum stiffness of about 40 Gurley in the machine
16 direction is sufficient to accomplish this purpose
17 and also to allow accurate sheet registration in the
18 forming of the stack 49 at the earlier stage of
19 manufacture illustrated in FIG. 3.

20 In accordance with the foregoing
21 processing conditions, the compositions of Examples
22 1, 2, 3 and 4 were coextruded, hot-stretched and
23 annealed to provide in-mold label films. The
24 properties of the films are reported below in Table
25 I.

26 TABLE I

27 EXAMPLE NUMBER	1	2	3	4	5	6
28 Thickness (mils)	4.0	4.0	4.0	4.5	4.0	4.0
29 Opacity (%)	85.0	10.5	10.5	10.5	85.0	9.7
30 Gurley Stiff. MD	80	45	80	115	80	65
31 (mg)	CD	35	20	35	50	35
32 Ten. Mod.	MD	285	145	270	270	285
33 (1000 psi)	CD	100	45	90	90	100
						75

1	Elong. (%)	MD	45	55	45	45	45	44
2		CD	275	800	500	500	275	925
3	Ten. Str.	MD	25.0	18.0	27.0	27.0	25.0	23.0
4	(1000 psi)	CD	2.5	2.2	3.3	3.3	2.5	3.5

5 Films having a composition in accordance
6 with Examples 1 through 6 have been used for in-mold
7 labelling of high density polyethylene containers.
8 As described below, the printing and label
9 converting processes are performed using
10 conventional paper label apparatus and techniques.

11 The labels were printed using typical
12 paper label printing presses, such as a Gallus
13 press, and UV curing inks. The films were pulled
14 through the press with a minimum force of about 20
15 pounds on a 6.5 inch wide web so as to impose a
16 loading of about 3 pounds per linear inch. Even
17 though UV curing inks are used, the ultraviolet
18 radiation source generates heat and the temperatures
19 in the cure portion of the press may range from
20 ambient (e.g. about 70 degrees F.) up to about 140
21 to 150 degrees F. The film of Example 3 was printed
22 using printing conditions similar to those used for
23 paper labels. The film displayed sufficient
24 dimensional stability to enable maintenance of
25 printing registration and did not excessively
26 stretch or elongate so as to otherwise prevent
27 processing.

28 The printed labels were sheeted using
29 conventional techniques as described above. The
30 antistat in the adhesive or base layer of the films
31 of Examples 1 through 4 effectively reduced and/or
32 eliminated static charge problems in the handling
33 and stacking of the cut film and labels. The over-

1 varnish used in the films of Examples 5 and 6
2 similarly reduced and/or eliminated static charge
3 problems.

4 The labels of Examples 1 through 6 were
5 applied to containers using both shuttle and rotary
6 blow-molding machines operating at typical
7 production rates. More particularly, labels were
8 applied using a Beckum BETM shuttle two-cavity blow-
9 molding machine operating at a rate of about 30
10 containers per minute and a Graham Engineering
11 rotary blow-molding machine operating at rate of
12 about 60 containers per minute. In these
13 applications, a low melt flow, high density
14 polyethylene resin, such as that sold by Quantum
15 Chemical under designation number 5602, was molded
16 at a parison temperature of 390 degrees F. as
17 measured at the barrel exit. The robotic arms
18 automatically disposed the labels in the mold
19 without multiple label feed and/or label folding
20 problems. The labels adhered to the bottles along a
21 substantially bubble free adhesion interface. Upon
22 squeezing or deforming of the labelled bottle, the
23 label adhered and conformed to the bottle without
24 forming unacceptable fold or crease lines.

25 Referring to Fig. 9, a modified label film
26 100 comprises two or more laminae of multiple or
27 single layers. The film 100 has at least one non-
28 coextruded lamina or layer. For clarity of
29 disclosure, corresponding layers of the label films
30 10 and 100 are similarly numbered by adding 100 to
31 the reference numerals for the latter.

32 The label film 100 includes a coextruded
33 multilayer film lamina 101 comprising a top face
34 layer 112, a central or core layer 116 and an

1 additional bottom layer 117. The bottom layer 117
2 may be omitted as discussed below. The bottom layer
3 117 of the multilayer film lamina 101 or, in the
4 absence of such bottom layer, the exposed surface of
5 the core layer 116, is secured to a non-coextruded
6 adhesive-containing second film lamina or base layer
7 114 by extrusion coating, hot roll coating or some
8 other suitable film combining technique with or
9 without a tie coat.

10 The coextruded multilayer film lamina 101
11 is prepared by coextrusion through multifeed
12 coextrusion die 18 and processed in a similar manner
13 as the label film 10 and substantially the same
14 processing apparatus is used as indicated in Fig.
15 10. The first film lamina 101 is cast "bottom layer
16 117 up" (i.e. with the bottom layer 117 on the upper
17 side and the layer 112 on the lower side).
18 Accordingly, the bottom layer 117 is conveniently
19 facing upwardly for combination with the second film
20 lamina or base layer 114. Of course, the first film
21 lamina 101 may be cast in an opposite orientation
22 and a transfer box (not shown) may be used to invert
23 the film as is well known in the art.

24 The second film lamina or base layer 114
25 may be combined with the first film lamina 101
26 before, during or after processing in the MDO unit
27 25. In Fig. 10, the base layer 114 is shown
28 extrusion coated onto the bottom layer 117 after
29 processing of the film 101 in the MDO unit 25.

30 The multilayer film lamina 101 is
31 stretched and stiffened in the machine direction in
32 the MDO unit 25 in a similar manner as described
33 above with respect to the film 10. However, the
34 processing in the MDO unit 25 is done at higher

1 temperatures in this embodiment to shorten the
2 processing time since the second lamina 114
3 containing the heat-activatable adhesive is
4 subsequently combined with the multilayer film
5 lamina 101. Accordingly, the following roll
6 temperatures are used: preheat rolls 26 and 28
7 (250°F), slow draw roll 31 (260°F), fast draw roll 32
8 (265°F), and annealing roll 36 (270°F). Thereafter,
9 the film 101 is cooled by contact with chill roll 38
10 which may be at a temperature of about 140°F. If the
11 second lamina 114 is combined with the multilayer
12 film lamina 101 prior to processing in the MDO unit
13 25, the above described time-temperature-direction
14 relationships may be employed to effect hot-
15 stretching and annealing without activating the
16 adhesive so as to result in sticking to the rolls.

17 As shown in Fig. 10, the second lamina or
18 base layer 114 is extrusion coated using die 121 to
19 cast the 100% solids polymeric charge of the second
20 lamina or base layer 114 onto the bottom layer 117
21 of multilayer first film lamina 101 to thereby
22 combine the second lamina with the hot-stretched and
23 annealed first film lamina. Following the combining
24 of the first and second laminae 101 and 114 to form
25 the label film 100, the film is cooled with suitable
26 chill rolls (not shown) disposed downstream of the
27 die 121. Thereafter, the film 100 is taken up on
28 the self-wound roll 139. Alternatively, the cooled
29 multilayer film lamina 101 exiting from the MDO unit
30 25 may be wound on the roll 139 and later coated or
31 otherwise combined with the second lamina or base
32 layer 114.

1 The uniaxial hot-stretching of the
2 multilayer film lamina 101 substantially increases
3 stiffness in the machine direction and leaves the
4 stock relatively flexible in the cross direction to
5 improve conformability with a container to which it
6 is to be applied, e.g. deformable plastic containers
7 which are flexed during use. Uniaxial hot-
8 stretching and annealing are also important in the
9 development of the tensile properties necessary to
10 withstand the mechanical and thermal stresses of
11 conventional paper printing processing. The useful
12 and preferred ranges of stiffness and tensile
13 properties noted above for the label film 10 are
14 applicable to the label film 100 as provided in
15 Example 7 by the hot-stretching and annealing of the
16 multilayer film lamina 101. It should be
17 appreciated that the second lamina or base layer 114
18 is relatively thin, and its omission from processing
19 in the MDO unit 25 does not prohibit or
20 significantly inhibit the development of the desired
21 stiffness and tensile properties of the label film
22 100.

23 The thickness of the label film 100 is
24 similar to that of the label film 10, and similar
25 layer thickness ratios are applicable. The
26 thickness of the bottom layer 117 of multilayer
27 lamina 101 is minimized to that necessary to prevent
28 migration of filler (e.g. titanium dioxide) from the
29 central or core layer 116. Thus, the extruded or
30 cast thickness of the layer 117 may be one mil or
31 less. In cases where it is not necessary to confine
32 a filler within the central layer, the layer 117 may
33 be omitted so as to provide a label film 100 having
34 a layer construction similar to that of the film 10.

1 The following example illustrates the use
2 of a coextruded multilayer film lamina of non-
3 adhesive layers which is subsequently combined with
4 a lamina comprising an adhesive layer as described
5 above with respect to the film 100. In this
6 example, the coextruded non-adhesive multilayer film
7 lamina is formed of layers having the following
8 compositions indicated by weight percentage.

<u>Example 7</u>		
10	Top	polypropylene homopolymer 49
11		ethylene-vinyl acetate copolymer 49
12		diatomaceous earth 2
13	Central	polypropylene homopolymer 44
14		high density polyethylene 20
15		ethylene-vinyl acetate copolymer 5
16		talc 26
17		titanium dioxide concentrate 5
18	Bottom	polypropylene homopolymer 89
19		ethylene-vinyl acetate copolymer 9
20		diatomaceous earth 2
21	The composition of the various layers in	
22	Example 7 is similar to those of Examples 1-6 in	
23	that olefin polymers and copolymers of olefin	
24	monomers with ethylenically unsaturated carboxylic	
25	acid or ethylenically unsaturated carboxylic acid	
26	ester comonomers such as ethylene-vinyl acetate	
27	copolymer are used. Herein, the central layer	
28	includes a high density polyethylene component	
29	comprising a fractional melt, film-grade resin	
30	material sold by Solvay Polymers, Inc. of Houston,	
31	Texas. An increased amount of polypropylene is used	
32	in the layer 117 for additional stiffness. The	
33	titanium dioxide filler in the core layer 116	

1 provides the desired opaque or white color. The
2 diatomaceous earth is an antiblocking agent.

3 The non-adhesive multilayer film lamina
4 was coextruded using the above described conditions.
5 The total thickness of the hot coextrudate was about
6 20 mils. The central or core layer forms the
7 majority of the thickness of the multilayer film
8 lamina, equal to about 80% thereof, and each of the
9 top and bottom layers equals about 10% of the total
10 thickness. The multilayer film lamina was hot-
11 stretched at a 5:1 ratio and annealed using the
12 above described conditions. The stretching of the
13 multilayer film lamina resulted in the expected
14 reduction in thickness and, accordingly, the
15 thickness of the multilayer film lamina was reduced
16 to about 4 mils.

17 A charge for the second lamina or base
18 layer was prepared solely of the above described
19 H.B. Fuller product number HM727 which is an EVA
20 based heat-activatable adhesive. The base layer was
21 extrusion coated onto the nonadhesive layers of the
22 multilayer film lamina after processing in the MDO
23 unit. The extruded thickness of the adhesive layer
24 was about 1.3 mils.

25 The resulting film in accordance with
26 Example 7 is well balanced with respect to heat-
27 shrinkability since the compositions of the various
28 layers are similar. The inclusion or omission of
29 the bottom layer of the first or multilayer film
30 lamina does not appear to affect the balance to a
31 detectable degree. The range of useful polymeric
32 constituents corresponds to that described above for
33 Examples 1-6.

1 The properties of the film 100 are
 2 reported below in Table II.

3 TABLE II

4	<u>EXAMPLE NUMBER</u>	7
5	Thickness (mils)	5.3
6	Opacity (%)	83.0
7	Gurley Stiff. MD	71
8	(mg) CD	55
9	Ten. Mod. MD	152.4
10	(1000 psi) CD	78.0
11	Elong. (%) MD	42
12	CD	230
13	Ten. Str. MD	11.1
14	(1000 psi) CD	2.1

16 Labels were prepared using the film of
 17 Example 7 and applied to containers during the
 18 molding process as in-mold labels in the same manner
 19 as in Examples 1-6. This film and resulting labels
 20 were found to perform in a satisfactory manner.

21 The following comparative examples
 22 illustrate the effects of various physical
 23 properties of the subject films in the label
 24 manufacture and application processes. In each of
 25 the following comparative examples, the compositions
 26 were formed into three layer films of the indicated
 27 total thickness and a 10/80/10 relative layer
 28 thickness ratio using the above described
 29 coextrusion, hot-stretching and annealing techniques
 30 to provide in-mold label films.

31 Comparative Example 1C

32	Top	polypropylene homopolymer	50
33		ethylene-vinyl acetate copolymer	50

1	Central	polypropylene homopolymer	30
2		high density polyethylene	33
3		ethylene-vinyl acetate copolymer	15
4		talc	15
5		titanium dioxide	7
6	Base	polypropylene homopolymer	50
7		ethylene-vinyl acetate copolymer	50

A 3.5 mil thick in-mold label film of the composition of Comparative Example 1C had a machine direction Gurley stiffness value in the range of from about 40 to about 48. Due to the relatively low Gurley stiffness values and the absence of an antistat component, the handling characteristics of this film were not entirely satisfactory during sheeting and the cut film did not uniformly stack since the relatively limp cut film pieces did not easily slide over each other and such movement was impeded by any static charges present. For similar reasons, labels formed from this film and used in a Graham Engineering rotary blow-molding machine in the above described manner did not dispense satisfactorily in that the label tended to fold and/or flap as it was placed in the mold. Further, the base layer failed to stick to a high density polyethylene container.

Comparative Example 2C was the same as Comparative Example 1C except that it had a total film thickness of 4 mils and the base layer composition was as follows: polypropylene homopolymer (50), ethylene-vinyl acetate copolymer (25), and heat activatable adhesive (25). The film of Comparative Example 2C had a Gurley value of about 60, and it satisfactorily sheeted and dispensed when processed in a manner similar to

1 Comparative Example 1C. However, there was a lack
2 of adhesion sufficient to affect label and container
3 conformability. The adhesion deficiencies are
4 indicated by bubbles along the adhesion interface
5 between the container and adjacent label base
6 surface. Comparative Example 3C included
7 modification of the base layer formulation to that
8 of Examples 1-3 herein and achievement of
9 satisfactory adhesion and static charge properties.
10 This film had a machine direction Gurley stiffness
11 value of about 60. The film displayed satisfactory
12 film processing characteristics and resulted in
13 labels having acceptable adhesion and conformability
14 properties when applied to high density polyethylene
15 containers. However, surface mottling possibly due
16 to water absorption by the talc was observed.

17 Comparative Example 4C comprised a
18 modification of the formulation of Comparative
19 Example 3C wherein polypropylene homopolymer was
20 substituted for the talc component. Surprisingly,
21 an additional increase in Gurley from about 60 to
22 about 80 was attained. This additional stiffness is
23 believed beneficial.

24 As shown in Tables I and II, uniaxial
25 stretching provides the films of Examples 1-7 with
26 dimensional stability to enable linerless processing
27 thereof. More particularly, the uniaxial stretching
28 techniques of the invention tend to stabilize the
29 film and facilitate linerless film processing as,
30 for example, label printing using conventional
31 printing presses as illustrated above. The
32 relatively low elongation values at break and the
33 high tensile modulus values in the machine direction
34 are believed to characterize acceptable dimensional

1 stability of the film correlating to the stretching
2 techniques of the invention.

3 Referring to Table III below, Comparative
4 Examples 5C and 6C comprise commercially available
5 in-mold label film. These films are believed to
6 comprise multilayer coextruded polyolefin
7 constructions of primarily medium density
8 polyethylene.

9 Table III

10	11	Comparative <u>Example Number</u>	5C	6C
12	Thickness (mils)		4.5	4.5
13	opacity (%)		12.5	78.0
14	Gurley Stiffness (mg)	MD	45	55
15		CD	55	60
16	Tensile Modulus (1000 psi)	MD	65	65
17		CD	70	75
18	Elongation (%)	MD	950	850
19		CD	1150	1050
20	Tensile Strength (1000 psi)	MD	3.2	2.6
21		CD	2.9	2.7

22 As indicated by the stiffness, tensile and
23 elongation properties, it is not believed that the
24 films of Comparative Examples 5C and 6C are hot-
25 stretched as in the present invention. However, the
26 films of Comparative Examples 5C and 6C may be
27 imparted with a limited degree of melt orientation
28 during the coextrusion process. The limited
29 orientation, if any, of these films results in
30 relatively low tensile modulus and elongation
31 properties in the machine direction which are
32 believed to be associated with the printing problems
33 encountered with such films. More particularly,
34 films in accordance with Comparative Examples 5C and

1 6C tend to elongate excessively during printing upon
2 application of press tension and temperature
3 conditions described above so as to prevent or
4 substantially inhibit maintenance of print
5 registration and film handling by conventional
6 printing techniques. Thus, it is believed that
7 linerless film processing of in-mold film using
8 conventional printing techniques requires a machine
9 direction tensile modulus greater than about 65,000
10 psi and/or an elongation at break less than about
11 850%.

12 While the invention has been illustrated
13 and described with respect to the in-mold labelling
14 of blow-molded containers, the invention may be
15 directly applied and its benefits may be directly
16 enjoyed in other thermal forming techniques such as
17 injection molding, thermoforming and sheet molding
18 compound forming when used to form a container or
19 substrate against a mold wall having a label or
20 other film (e.g. decorative film) positioned
21 thereon.

22 In the application of the time-
23 temperature-direction conditions of the invention to
24 labels or other film stock, it should be understood
25 that the heat energy for hot-stretching and
26 annealing may be provided by conventional film
27 processing apparatus such as heated rolls as
28 described above (with oil or radio frequency
29 heating) as well as infrared heat sources such as
30 infrared heat lamps, and combinations thereof, as
31 are well known in the art. The time-temperature-
32 direction conditions of the invention may be used in
33 connection with all such heating techniques.

1 It should be evident that this disclosure
2 is by way of example, and that various changes may
3 be made by adding, modifying or eliminating details
4 without departing from the fair scope of the
5 teaching contained in this disclosure. The
6 invention therefore is not limited to particular
7 details of this disclosure except to the extent that
8 the following claims are necessarily so limited.

WHAT IS CLAIMED IS:

1 1. An in-mold labelling method comprising
2 the steps of coextruding a plurality of at least two
3 charges of film-forming resin, coextruding said
4 charges to thereby form a construction in the form
5 of a multilayer extrudate having a face side and a
6 back side, preselecting said charges to provide a
7 printable face on said face side and a heat-
8 activated adhesive at said back side, hot-stretching
9 and annealing said extrudate to thereby provide a
10 machine direction stiffness differential and enhance
11 the dimensional stability of the free-film
12 extrudate, printing the face side of the free-film
13 extrudate and exposing the extrudate to a drying
14 agent such as heat or U.V. to dry the ink, die-
15 cutting the free-film extrudate as individual
16 labels, and sequentially deploying the labels on a
17 molding surface of a mold for bonding onto
18 successive workpieces as said workpieces and said
19 molding surface are brought together in the presence
20 of heat whereby said adhesive is activated and
21 contacted with said workpieces.

1 2. A method as in claim 1, said extrudate
2 having an annealing temperature above the
3 temperature at which said adhesive is activated,
4 said step of hot-stretching and annealing including
5 passing said extrudate across heating and cooling
6 means including roll means contacting said extrudate
7 to thereby impart heat to and remove heat from said
8 extrudate under time-temperature-direction
9 conditions established by line speed, temperature of

10 said heating and cooling means and side of heat
11 contact, said step of hot-stretching and annealing
12 further including controlling said time-temperature-
13 direction conditions to heat at least a majority of
14 the thickness of the extrudate to above its
15 annealing temperature following stretching without
16 sticking of said adhesive to said roll means,
17 despite said annealing temperature being above the
18 temperature at which said adhesive is activated.

1 3. A method as in claim 2, wherein said
2 heating and cooling means comprise a series of
3 relatively hot and cool rolls through which said
4 extrudate is trained.

1 4. A method as in claim 2, said step of
2 hot-stretching and annealing including heating the
3 face side of said extrudate with said heating and
4 cooling means heated to a temperature above the
5 temperature of activation of the adhesive so as to
6 impart heat from said heating and cooling means to
7 the construction without flowing said heat through
8 said adhesive.

1 5. A method as in claim 1, said extrudate
2 having a softening temperature above the temperature
3 at which said adhesive is activated, said extrudate
4 having an annealing temperature also above the
5 temperature at which said adhesive is activated,
6 said step of hot-stretching and annealing including
7 passing said extrudate across heating and cooling

8 means including roll means contacting said extrudate
9 to thereby impart heat to and remove heat from said
10 extrudate under time-temperature-direction
11 conditions established by line speed, temperature of
12 said heating and cooling means, and side of heat
13 contact, said step of hot-stretching and annealing
14 further including controlling said time-temperature-
15 direction conditions to heat at least a majority of
16 the thickness of the extrudate to above its
17 softening temperature prior to stretching without
18 sticking of said adhesive to said roll means, and
19 heating at least a majority of the thickness of the
20 extrudate to above its annealing temperature
21 following stretching without sticking of said
22 adhesive to said roll means, despite both said
23 softening temperature and said annealing temperature
24 being above the temperature at which said adhesive
25 is activated.

1 6. A method as in claim 5, said step of
2 hot-stretching and annealing including heating the
3 face side of said extrudate with said heating and
4 cooling means heated to a temperature above the
5 temperature of activation of the adhesive so as to
6 impart heat from said heating and cooling means to
7 the construction without flowing said heat through
8 said adhesive.

1 7. A method as in claims 2, 4 or 5,
2 wherein said heating and cooling means comprise a
3 series of relatively hot and cool rolls through
4 which said extrudate is trained and include an

5 annealing roll for contacting the face side of said
6 extrudate..

1 8. A method as in claim 1, wherein said
2 mold is a blow mold and said workpieces are blown
3 and expanded against said molding surface.

1 9. A method as in claim 1, said
2 preselecting step including preselecting said
3 charges to contain major proportions of like
4 materials to thereby balance the heat-shrinkability
5 at each side of said extrudate to a sufficient
6 extent to limit curling of the extrudate following
7 hot-stretching.

1 10. A method as in claims 1 or 9, said
2 preselecting step including providing an antistat
3 agent in the charge for the layer which includes the
4 heat-activatable adhesive.

1 11. A method as in claim 1, including
2 hot-stretching and annealing said extrudate to
3 provide said extrudate with a machine direction
4 stiffness differential.

1 12. A method as in claim 1, including
2 uniaxially hot-stretching said extrudate at a
3 stretch ratio in the range of from about 2 to 1 to
4 about 8 to 1.

1 13. A method as in claim 1, including
2 uniaxially hot-stretching said extrudate at a
3 stretch ratio in the range of from about 4 to 1 to
4 about 6 to 1.

1 14. A method as in claim 1, including
2 between the steps of hot-stretching and printing,
3 self-rolling said extrudate, transporting said self-
4 rolled extrudate, and unrolling said self-rolled
5 extrudate.

1 15. A method as in claim 1, wherein said
2 stretched and annealed free-film extrudate has a
3 machine direction Gurley stiffness value in the
4 range of from about 40 to about 130 and a cross
5 direction Gurley stiffness value in the range of
6 from about 20 to about 65.

1 16. A method as in claim 1, wherein said
2 stretched and annealed free-film extrudate has a
3 tensile modulus value greater than about 65,000 psi.

1 17. A method as in claims 1 or 16,
2 wherein said stretched and annealed free-film
3 extrudate has a machine direction Gurley stiffness
4 value greater than about 45.

1 18. A method as in claim 17, wherein said
2 stretched and annealed free-film extrudate has a
3 machine direction elongation at break less than
4 about 850%.

1 19. A method as in claim 18, wherein said
2 stretched and annealed free-film extrudate has a
3 sufficiently high tensile modulus and stiffness
4 value in the machine direction to enable it to
5 withstand the mechanical and thermal stresses of
6 conventional printing processes including film
7 tension loads of about three pounds per linear inch
8 of film width at temperatures ranging from about 70
9 degrees F. to about 150 degrees F.

1 20. A method as in claim 1, in which said
2 stretched and annealed free-film extrudate has a
3 machine direction Gurley stiffness value greater
4 than its cross direction Gurley stiffness value and
5 a machine direction elongation at break less than
6 about 850%.

1 21. An in-mold labelling method
2 comprising, the steps of combining a plurality of at
3 least two laminae of film-forming resin to form a
4 label film having a face side and a back side with a
5 top layer at said face side and a base layer at said
6 back side, and either before, during or after said
7 combining step, hot-stretching and annealing said
8 first lamina to thereby provide a machine direction
9 stiffness differential and enhance the dimensional
10 stability of said label film which is to be, is
11 being, or has been formed, and before all the
12 aforesaid steps, preselecting the material for said
13 top layer to provide a printable face at said face
14 side of said label film and preselecting the
15 material for said base layer to provide a heat-
16 activated adhesive at said back side of said label
17 film, and following said combining, hot-stretching
18 and annealing steps, printing the face side of the
19 film and exposing the film to a drying agent such as
20 heat or U.V. to dry the ink, die-cutting the film as
21 individual labels, and sequentially deploying the
22 labels on a molding surface of a mold for bonding
23 onto successive workpieces as said workpieces and
24 said molding surface are brought together in the
25 presence of heat whereby said adhesive is activated
26 and contacted with said workpieces.

1 22. A method as in claim 21, wherein the
2 step of combining said plurality of at least two
3 laminae includes coextruding a plurality of at least
4 two charges of film-forming resin to thereby form
5 said first lamina and sec nd lamina as a
6 construction in the form of a multilay r extrudate.

1 23. A method as in claim 22, wherein the
2 coextruding step includes coextruding a third charge
3 intermediate said first and second charges to form
4 said multilayer coextrudate with a core or central
5 layer intermediate said printable face and adhesive.

1 24. A method as in claims 21, 22 or 23,
2 said film having an annealing temperature above the
3 temperature at which said adhesive is activated,
4 said step of hot-stretching and annealing including
5 passing said film across heating and cooling means
6 including roll means contacting said film to thereby
7 impart heat to and remove heat from said film under
8 time-temperature-direction conditions established by
9 line speed, temperature of said heating and cooling
10 means, and side of heat contact, said step of hot-
11 stretching and annealing further including
12 controlling said time-temperature-direction
13 conditions to heat at least a majority of the
14 thickness of the film to above its annealing
15 temperature following stretching without sticking of
16 said adhesive to said roll means, despite said
17 annealing temperature being above the temperature at
18 which said adhesive is activated.

1 25. A method as in claim 24, wherein said
2 heating and cooling means comprise a series of
3 relatively hot and cool rolls through which said
4 extrudate is trained.

1 26. A method as in claim 21, said step of
2 hot-stretching and annealing including heating the
3 face side of said film with said heating and cooling
4 means heated to a temperature above the temperature
5 of activation of the adhesive so as to impart heat
6 from said heating and cooling means to the
7 construction without flowing said heat through said
8 adhesive.

1 27. A method as in claim 26, wherein said
2 heating and cooling means comprise a series of
3 relatively hot and cool rolls which contact said
4 extrudate and include an annealing roll for
5 contacting the face side of said extrudate.

1 28. A method as in claims 21, 22 or 23,
2 said film having a softening temperature above the
3 temperature at which said adhesive is activated,
4 said film having an annealing temperature also above
5 the temperature at which said adhesive is activated,
6 said step of hot-stretching and annealing including
7 passing said film across heating and cooling means
8 including roll means to contact said film to thereby
9 impart heat to and remove heat from said film under
10 time-temperature-direction conditions established by
11 lin speed, heating means temperature, and side of

12 heat contact, said step of hot-stretching and
13 annealing further including controlling said time-
14 temperature-direction conditions to heat at least a
15 majority of the thickness of the film to above its
16 softening temperature prior to stretching without
17 sticking of said adhesive to said roll means, and
18 heating at least a majority of the thickness of the
19 film to above its annealing temperature following
20 stretching without sticking of said adhesive to said
21 roll means, despite both said softening temperature
22 and said annealing temperature being above the
23 temperature at which said adhesive is activated.

1 29. A method as in claim 28, wherein said
2 heating and cooling means comprise a series of
3 relatively hot and cool rolls which contact said
4 extrudate and include an annealing roll for
5 contacting the face side of said extrudate.

1 30. A method as in claim 21, wherein said
2 mold is a blow mold and said workpieces are blown
3 and expanded against said molding surface.

1 31. A method as in claim 28, said step of
2 hot-stretching and annealing including heating the
3 face side of said film with said heating and cooling
4 means heated to a temperature above the temperature
5 of activation of the adhesive so as to impart heat
6 from said heating and cooling means to the
7 construction without flowing said heat through said
8 adhesive.

1 32. A method as in claim 21, wherein said
2 combing and hot-stretching and annealing steps are
3 sequentially performed as said label film is
4 continuously produced.

1 33. A method as in claim 21, said
2 preselecting step including preselecting said
3 charges to contain major proportions of like
4 materials to thereby balance the heat-shrinkability
5 at each side of said label film to a sufficient
6 extent to limit curling of the film following hot-
7 stretching.

1 34. A method as in claim 21, including
2 uniaxially hot-stretching said first lamina at a
3 stretch ratio in the range of from about 2 to 1 to
4 about 8 to 1.

1 35. A method as in claim 21, including
2 between the steps of hot-stretching and printing,
3 self-rolling said label film, transporting said
4 self-rolled label film, and unrolling said self-
5 rolled label film.

1 36. A method as in claim 21, wherein said
2 label film has a machine direction Gurley stiffness
3 value in the range of from about 40 to about 130 and
4 a cross direction Gurley stiffness value in the
5 range of from about 20 to about 65.

1 37. A method as in claim 21, wherein said
2 label film has a tensile modulus value greater than
3 about 65,000 psi and a machine direction elongation
4 at break less than about 850%.

1 38. A method as in claim 37, wherein said
2 label film has a sufficiently high tensile modulus
3 and stiffness value in the machine direction to
4 enable it to withstand the mechanical and thermal
5 stresses of conventional printing processes
6 including film tension loads of about three pounds
7 per linear inch of film width at temperatures
8 ranging from about 70 degrees F. to about 150
9 degrees F.

1 39. A method as in claim 21, wherein said
2 base layer includes an antistat agent.

1 40. An oriented polymeric in-mold label
2 film comprising first and second laminae which
3 provide said film with a top layer at a face side of
4 the film and a base layer at a back side of the
5 film, said top layer being ink-printable and said
6 base layer comprising a heat-activatable adhesive,
7 at least said first lamina being hot-stretched and
8 annealed, said first and second laminae being
9 combined to form said film as a linerless self-wound
10 film, the heat shrinkability at each side of the
11 film being balanced to a sufficient extent to limit
12 curling of the film, said wound film being
13 unwindable and processable, as a free self-
14 supporting dimensionally stable film, through a
15 printing press, heat-generating ink drying means,
16 and die-cutting means.

1 41. A film as in claim 40, wherein said
2 top layer and said base layer each contain a major
3 proportion comprising a blend of an olefin polymer
4 and a copolymer of an olefin monomer with an
5 ethylenically unsaturated carboxylic acid or an
6 ethylenically unsaturated carboxylic acid ester
7 comonomer.

1 42. A film as in claim 40, wherein said
2 top layer and said base layer each contain a major
3 proportion comprising a blend of a polypropylene
4 polymer and a copolymer of an ethylene monomer with
5 an ethylenically unsaturated carboxylic acid or an
6 ethylenically unsaturated carboxylic acid ester
7 comonomer.

1 43. A film as in claim 40, wherein said
2 top layer and said base layer each contain more than
3 50% by weight of a blend of a polypropylene polymer
4 and an ethylene-vinyl acetate copolymer.

1 44. A film as in claim 40, wherein said
2 film has a machine direction Gurley stiffness value
3 in the range of from about 40 to about 130 and a
4 cross direction Gurley stiffness value in the range
5 of from about 20 to about 65.

1 45. A film as in claim 40, wherein said
2 film has a machine direction tensile modulus value
3 greater than about 65,000 psi.

1 46. A film as in claims 40 or 45, wherein
2 said film has a machine direction Gurley stiffness
3 value greater than about 40.

1 47. A film as in claim 46, wherein said
2 film has a machine direction elongation at break
3 less than about 850%.

1 48. A film as in claim 47, wherein said
2 film has a sufficiently high tensile modulus and
3 stiffness value in the machine direction to enable
4 it to withstand the mechanical and thermal stresses
5 of conventional printing processes including film
6 tension loads of about three pounds per linear inch
7 of film width at temperatures ranging from about 70
8 degrees F. to about 150 degrees F.

1 49. A film as in claim 40, wherein said
2 base layer includes an antistat agent.

1 50. A blow molded plastic container
2 having an in-mold label formed of a film in
3 accordance with claim 40.

1 51. A container as in claim 50, wherein
2 said container is formed of a polyolefin and said
3 top layer and said base layer of said film each
4 contain a major proportion comprising a blend of an
5 olefin polymer and a copolymer of an olefin monomer
6 with an ethylenically unsaturated carboxylic acid or
7 an ethylenically unsaturated carboxylic acid ester
8 comonomer.

AMENDED CLAIMS

[received by the International Bureau on 10 February 1993 (10.02.93);
original claims unchanged; new claims 52 and 53 added; (1 page)]

52. An in-mold labelling method comprising the steps of coextruding a plurality of at least two charges of film-forming resin, coextruding said charges to thereby form a construction in the form of a multilayer extrudate having a face side and a back side, preselecting said charges to provide a printable face on said face side and a heat-activated adhesive at said back side, uniaxially hot-stretching and annealing said extrudate to thereby enhance the dimensional stability of the free-film extrudate, printing the face side of the free-film extrudate and exposing the extrudate to a drying agent such as heat to dry the ink, die-cutting the free-film extrudate as individual labels, and sequentially deploying the labels on a molding surfaces of a blow mold for bonding onto successive hot workpieces as said workpieces are blown and expanded against said molding surface and against said label whereby contact by said hot workpieces activates said adhesive.

53. An in-mold labelling method comprising the steps of coextruding a plurality of at least three charges of film-forming resin, coextruding said charges to thereby form a construction in the form of a multilayer extrudate having a face layer, a back layer, and a core layer, preselecting said charges to provide a printable face at the face of said face layer and a heat activated adhesive at said back layer, uniaxially hot-stretching and annealing said extrudate to thereby enhance the dimensional stability of the free-film extrudate, printing said face of the face layer, die-cutting the free-film extrudate as individual labels, and sequentially deploying the labels on a molding surface of a blow mold for bonding onto successive hot workpieces as said workpieces are blown and expanded against said molding surface and against said label whereby contact by said hot workpieces activates said adhesive.

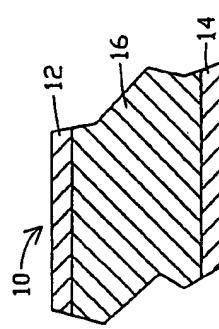


FIG. 1

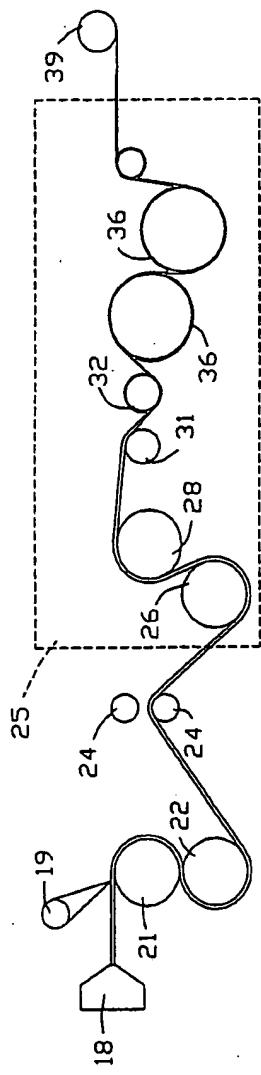


FIG. 2

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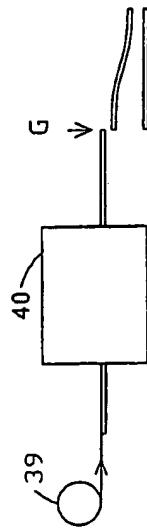


FIG. 3

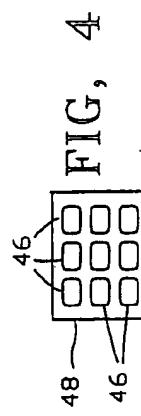


FIG. 4

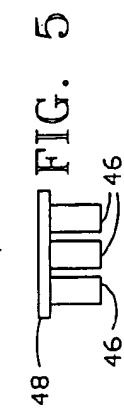


FIG. 5

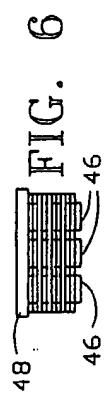


FIG. 6

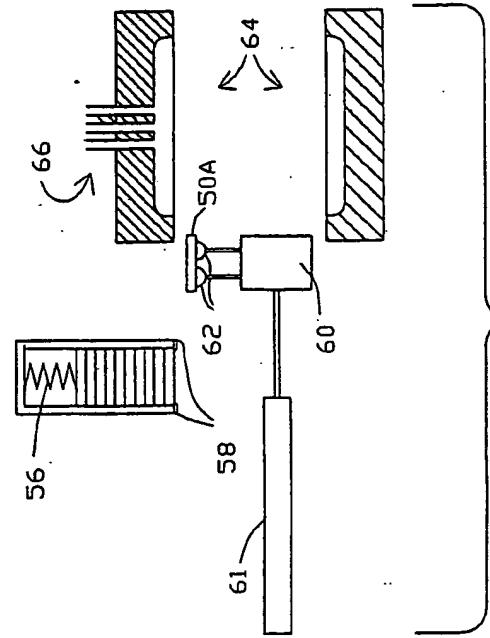


FIG. 7



FIG. 8

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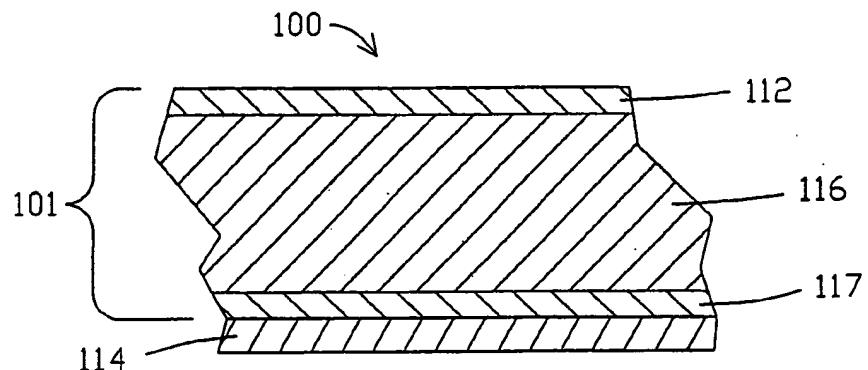


FIG. 9

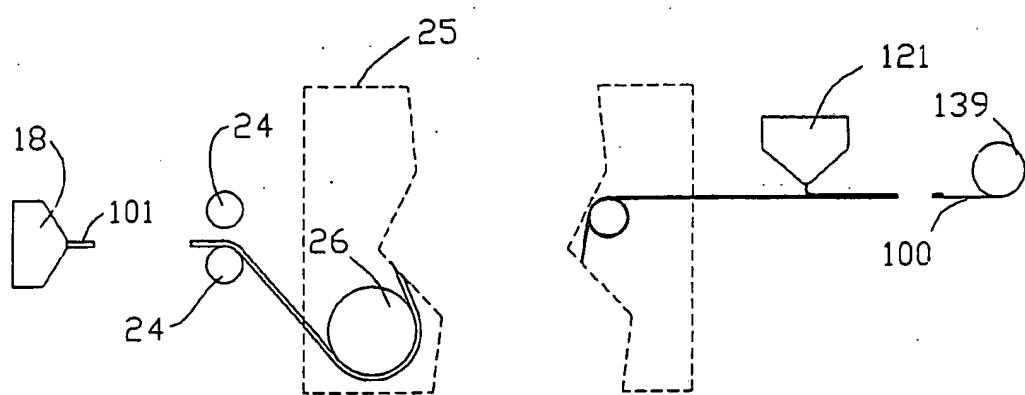


FIG. 10

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US92/07628**A. CLASSIFICATION OF SUBJECT MATTER**

IPC(5) :B29C 49/24, B29C 65/02, B29C 65/10

US CL :Please See Extra Sheet.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. :

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

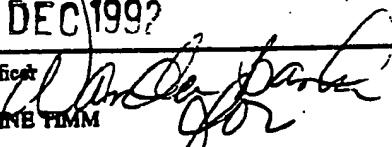
C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP,A, 2-217,223 (OjiYuka Goseishi KK) 30 August 1990 (Entire document).	1,8-23, 30,32-39
Y	US,A, 4,837,075 (Dudley) 06 June 1989 (Entire document).	1,8-23,30, 32-40,44-50
Y	US,A, 4,886,698 (Perdy) 12 December 1989 (col. 3, line 45 to column 4, line 68).	1,8-23,30, 32-39
Y	US,A, 4,501,797 (Super et al) 26 February 1985 (Entire document).	2-7,24-29,31,40 44-49
Y	US,A, 5,026,592 (Janocha) 25 June 1991.(entire document).	40,44-50
Y,P	US,A, 5,126,197 (Shinkel et al) 30 June 1992 (Column 1, line 52 to column 2, line 35).	41-42,51
A	US,A, 4,395,115 (Yoshii et al.) 12 July 1983.	1-51

 Further documents are listed in the continuation of Box C. See patent family annex.

* Special categories of cited documents:	"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
A document defining the general state of the art which is not considered to be part of particular relevance	"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
E earlier document published on or after the international filing date	"Y"	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
L document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"&"	document member of the same patent family
O document referring to an oral disclosure, use, exhibition or other means		
P document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search	Date of mailing of the international search report
12 NOVEMBER 1992	11 DEC 1992

Name and mailing address of the ISA/ Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. NOT APPLICABLE	Authorized officer:  CATHERINE TIMM Telephone No. (703) 308-3830
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INTERNATIONAL SEARCH REPORT

International application No.
PCT/US92/07628

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US,A, "Opticite Label films", Dow Chemical.	1-51
A	US,A, 4,986,866 (Ohba et al.) 22 January 1991.	1-51
A	US,A, 4,601,926 (Jabarin et al) 22 July 1986.	1-51
A	US,A, 4,904,324 (Heider) 27 February 1990.	1-51
A	CA,A, 2,012,357 (Kinoshita et al.) 17 September 1990.	1-51
A	US,A, 4,883,697 (Dornbush et al.) 28 November 1989.	1-51
A	US,A, 4,892,779 (Leatherman et al.) 9 January 1990.	1-51
A,P	US,A, 5,073,435 (Eyraud et al.) 17 December 1991.	1-51

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US92/07628

A. CLASSIFICATION OF SUBJECT MATTER:

US CL :

264/132,171,210.5,235.6,346,509;156/244.11,244.16,244.24;
428/35.7,359.9,343,347,349,354,483,520